Report IBM N00014-92-C-0193 Final

High-Temperature Superconducting Multi-Level Materials and Device Development and Device Physics

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19 January 1998

Final Report covering Sept. 30, 1992 to Sept. 30, 1995

Prepared for:

Office of Naval Research
Department of the Navy
800 North Quincy Street
Arlington, VA 22217

REPORT DOCUMENTATION PAGE

1a. REPORT SECURITY CLASSIFICATION
Unclassified

1b. RESTRICTIVE MARKINGS
N/A

2a. SECURITY CLASSIFICATION AUTHORITY
N/A

2b. DECLASSIFICATION/DOWNGRADING SCHEDULE
N/A

3. DISTRIBUTION/AVAILABILITY OF REPORT
~~Permitted for any purposed of the U.S. Government~~

4. PERFORMING ORGANIZATION REPORT NUMBER(S)
IBM N00014-92-C-0193 Final

5. MONITORING ORGANIZATION REPORT NUMBER(S)

6a. NAME OF PERFORMING ORGANIZATION
IBM Thomas J. Watson Research Center

6b. OFFICE SYMBOL (If applicable)

7a. NAME OF MONITORING ORGANIZATION
Office of Naval Research

6c. ADDRESS (City, State and ZIP Code)
P.O. Box 218, Yorktown Heights, NY 10598

7b. ADDRESS (City, State and ZIP Code)
800 North Quincy Street
Arlington, VA 22217

8a. NAME OF FUNDING/SPONSORING ORGANIZATION
Office of Naval Research

8b. OFFICE SYMBOL (If applicable)

9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER
N00014-85-C-0361

8c. ADDRESS (City, State and ZIP Code)
Department of the Navy
800 N. Quincy St., Arlington, VA 22217-5000

10. SOURCE OF FUNDING NOS.

PROGRAM ELEMENT NO.	PROJECT NO.	TASK NO.	WORK UNIT NO.

11. TITLE (Include Security Classification)
High Tc Materials, Devices (unclassified)

12. PERSONAL AUTHOR(S)
Gallagher, William J.; Koch, Roger H.

13a. TYPE OF REPORT
Final Tech. Report

13b. TIME COVERED
FROM 9/30/92 TO 9/30/95

14. DATE OF REPORT (Yr., Mo., Day)
January 19, 1998

15. PAGE COUNT
9

16. SUPPLEMENTARY NOTATION

17. COSATI CODES

FIELD	GROUP	SUB. GR.

18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number)
High-Temperature Superconductivity
Superconducting Quantum Interface Devices (SQUIDS)

19. ABSTRACT (Continue on reverse if necessary and identify by block number)

A three year research effort aimed at exploring the potential of the new high-Tc superconductors for SQUID applications is described. Over the course of the contract advances in multilayer materials and junction devices were made, particularly in the reduction of hysteresis and in the demonstration in high-Tc technology of a three squid gradiometer. The latter approach is particularly well suited for further development and application for Naval gradiometer purposes.

20. DISTRIBUTION/AVAILABILITY OF ABSTRACT
UNCLASSIFIED/UNLIMITED ☒ SAME AS RPT. ☐ DTIC USERS ☐

21. ABSTRACT SECURITY CLASSIFICATION
Unclassified

22a. NAME OF RESPONSIBLE INDIVIDUAL
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(914) 945-2483

22c. OFFICE SYMBOL

DD FORM 1473, 83 APR EDITION OF 1 JAN 73 IS OBSOLETE.

Final report

High-Tempature Superconducting Multi-level Materials and Device Development and Device Physics

William J. Gallagher, Roger Koch, Principal Investigators

Contract N00014-92-C-0193

Contract period: Sept. 30, 1992 to Sept. 30, 1995

1. Technical Objectives

The goals of contract N00014-92-C-0193 were: (1) to understand and improve high-Tc junctions for future gradiometers, particularly by lowering white and 1/f noise and by developing processes and device structures to make practical gradiometer SQUIDs configurations; (2) to understand magnetic hysteresis in SQUIDs and gradiometers and to devise and implement materials, process, and structural changes to lower hysteresis; (3) to scale up the size of high-Tc multilevel pick-up coils by an order of magnitude and combine these with optimized flux focusing transformer primaries; (4) to demonstrate a novel three SQUID gradiometer approach that could favorably impact the demands on increasing coil size and lowering hysteresis, and ease the fabrication requirements for naval gradiometers.

2. Methodology

The approach taken involved four basic thrusts: (1) investigation of high-Tc junction materials and structures paying particular attention to improved 1/f noise, yield, and reliability; (2) the use of the best available 77 K films and device structures to fabricate and characterize exploratory SQUIDs, magnetometers, and gradiometers paying particular attention to key practical issues such as hysteresis, noise in field, and RFI-immunity; (3) fabrication of high quality superconducting and insulating films on scales up to 2 inches as components of multilevel structures; (4) characterization of devices fabricated both with regard to their device performance and the underlying device physics and feedback these results to make improved devices.

3. Accomplishments

Thousand-Fold Improved Hysteresis in 77 K SQUIDs

Magnetic hysteresis in a thin film SQUID manifests itself as a shift of the SQUID's voltage-field transfer function along the field axis after a brief exposure to an intense (e.g. 1 to 10 G) magnetic

field. It is an undesirable feature for applications in which either the absolute value of field is to be determined, or small gradients of field are to be measured in significant background fields. In 1993, utilizing understanding gained from hysteresis measurements and modelling and improved high-Tc device processing, we made hysteresis improvements in 2 to 4 Gauss fields at 77 K of three orders of magnitude and obtained clear evidence for a threshold field that was predicted by the model we earlier developed for hysteresis. The model had indicated that edge current densities were low in our samples, and that an image force at the edge should give rise to a threshold field. Processing steps we implemented to give the improved the edge quality were a lowering of the duty cycle used for ion milling and a post processing treatment with an oxygen plasma. Inferred edge current densities, consistent with both the observed hysteresis and the threshold field, range from 1 to 3 x 10^6 A/cm^2 at 77 K.

Improved High-Tc Three SQUID Gradiometer

In 1993 we constructed an improved 77 K Three SQUID Gradiometer (TSG) and operated it in an unshielded laboratory environment with a gradient sensitivity sensitivity of 6 pT/m Hz.. This was a white noise level almost as good as the low-Tc naval gradiometer that the Navy was preparing to develop and deploy. The two main improvements in the current TSG were the incorporation of flux focusing structures on the upper and lower sensors and the use of lower noise step edge SQUIDs. The noise improvement was 5-fold and was due to use of the improved step edge junction SQUIDs developed over the last year. The use of press-on 3mm x 3mm washers resulted in a net responsivity improvement of 15X. On the other hand, this TSG had a shorter baseline of 10 cm, which was 3 times shorted than that of the first TSG's, developed in 1992 under a prior contract. This combination of factors accounted for the 23-fold improvement over the earlier 77 K TSG. This 1993 TSG was used to look at a number of performance related issues of importance for future designs. Among these are alignment and balance issues, RFI interference issues, and noise increases associated with cooling and operation in ambient fields.

Improved 1/f Noise in 77 K SQUIDs with Step-Edge Junctions

In 1993 we applied bias-reversing techniques to lower by up to 30X the 1/f device noise in our low-noise step-edge junction SQUIDs. The result was been the demonstration of an 80 pH device with a noise level of 19 $\mu\Phi_0/\sqrt{\text{Hz}}(10^{-29}$ J/Hz) at 1 Hz.

Three SQUID gradiometer

During the FY93-94 contract period, we reduced to practice for the first time a recently IBM invented (by P.I. R.H. Koch) Three SQUID Gradiometer (TSG). The TSG uses three SQUIDs, with or without magnetic field pickup coils. One SQUID is designated the *reference SQUID or magnetometer*. This SQUID operates with the usual feedback loop. Hence, the SQUID operates as a null detector, i.e. any change in external magnetic flux applied to this SQUID is cancelled by an equal and opposite flux generated by the feedback coil. The feedback current, however, is directed not only to the reference SQUID, but also to the two other SQUID magnetometers. We employ a special coil, the *triple coil*, to apply this common feedback signal. The triple coil, made from copper wire, is wound such that the current through it generates the same vector

magnetic field at each of the three SQUID locations. With this arrangement, a uniform magnetic field change applied along the baseline of the gradiometer is nulled at the location of all three SQUIDs. When a non-uniform field is applied, the two sensor SQUIDs experience only the difference in the field magnitude between the sensor and reference SQUID locations. The measured average gradient is the difference in magnetic fields indicated by the two feedback loops of the two sensor SQUIDs divided by the distance between the two SQUIDs. In our initial demonstrations, which was for a TSG with a 28 cm baseline, we measured gradient sensitivities of 10^{-12} and 10^{-10} T/m√Hz for TSG's made from bare low-Tc and high-Tc SQUIDs, respectively. Following this initial demonstration, we have been constructing improved TSGs. A TSG with a 10 cm baseline and lower noise SQUIDs with flux focussing washers pressed on to increase their field sensitivities had a gradient sensitivity of 3 pT/m Hz at high frequencies at 77 K. This white noise level was almost as good as the (albeit flat to 0.1 Hz) noise level of a recent demonstration 4.2 K gradiometer evaluated by the Navy. The current TSG has been used to look at a number of performance related issues of importance for future designs. Among these are alignment and balance issues, RFI interference issues, and noise increases associated with cooling and operation in ambient fields.

The effects of radio frequency radiation on the dc SQUID

SQUID are usually operated in rf shielded environments. For applications in which SQUIDs are moved, eddy currents induced in the shielding materials are major noise sources. In order to determine how we might avoid the use of rf shields, we studied the effects of rf radiation on SQUIDs. Simulations were used to show how the SQUID transfer characteristic is distorted by radio frequency interference (RFI). How this affects three commonly used SQUID modulation methods was examined. The results explained experiments in which we observed the bias current reversing readout method to be the least susceptible to RFI. The commonly seen increase in the low frequency flux power noise spectrum of the dc SQUID in unshielded environments was also explained.

Reliable high-Tc SQUIDs with performance matching the RSJ model

In conjunction with needs in other programs made possible by earlier results in the high-Tc SQUID research development effort, a significant focus in 1994-95 was on reproducibly and reliably achieving high-sensitivity magnetometers of order 100-200 fT/√Hz at ~1 Hz. This level of performance was consistent with intermediate term application needs and with the use of single-level high-Tc magnetometers in the three SQUID gradiometer configuration. Key to improving reliability was resolving handling issues (moisture exposure, electrostatic discharge) encountered in instrumental applications of our devices. In joint work with our SQUID commercialization partner, Quantum Magnetics, we developed a teflon passivation coating and incorporated this into a packaging scheme that has greatly improved device survivability in instrumental applications. Simultaneous with this and with more careful definition and practice of our fabrication procedures, the responsivity of our devices has improved and now for many devices matches the predictions of the Resistively Shunted Junction (RSJ) model. This is true for both bicrystal and step edge devices. Up until this time, similar comparisons between theory and experiment always found a reduction in the performance (i.e the SQUID transfer function)

in the actual devices when compared to numerical predictions of the SQUID response. The reduction of the SQUID response was approximately proportional to the SQUID critical current - resistance product. This was true for any SQUIDs fabricated by any group world wide. Our new series of SQUIDs do not show this reduction in performance, with over 50 devices being within the expected factor of two of predictions. The exact reason why the earlier devices were deficient is not 100% clear, but we believe that the performance was downgraded during the lithographic patterning process and, because the earlier devices were not passivated, during the testing process.

New approach to low-noise flux-gate magnetometers

In conjunction with our work on three-magnetometer gradiometers which incorporate a flux-gate reference magnetometer and two SQUID magnetometers, it is desirable to have as absolute reference magnetometer with as low of noise as possible. Noise at low frequency is associated with domain motion and know as Barkhausen noise. We are investigating a single domain approach to a flux-gate to completely eliminate this noise source. At the present time we have achieved a noise level of 5-10 pT/√Hz at 1 Hz, limited (we believe) by amplifier noise using a FeNiMo alloy with the trade name Hipernom, which matches the noise level of the best commercial fluxgates in our laboratory. Future work wwas planned to establish the intrinsic noise limits of the current approach to flux-gate sensors, and indicate their proper role in future three-magnetometer gradiometers. This work formed the basis of a follow-on contract.

4. Significance

All the high-Tc technology in this contract was developed to the point it was adequate for naval device demonstrations that have given a reasonable indication that adequate performance would be achieved. Our successful demonstrations of three SQUID gradiometers give a further indication that 5 cm high-Tc technology may be all that is required for the long run -- i.e. larger superconducting coil structures will not be needed for making conventional gradiometers. To be sure, however, technology improvements were still needed. The demonstrations to date indicate that further improvements by one to two orders of magnitude are needed in the area of junction 1/f noise. There was reason for optimism in this regard because of the steady low frequency noise advances that were achieved in this time frame as high-Tc thin film technology has matured. Realizing further low frequency noise improvements was a primary objective of a proposed follow-on contract.

In addition, further optimization of designs and processes for multilevel flux-focusing transformers was needed to fully achieve performance projections. So far the multilevel technology for such coils, particularly at 5 cm, has lagged the single level technology. High-yield, optimized multilevel technology seems feasible with further technology work.

The white noise level now demonstrated in our 77 K TSG was almost as good as the low-Tc gradiometer that IBM FSC developed based on Quantum Design 4.2 K SQUIDs and Nb wire-wound pick-up coils. Complete proof of viability of 77 K TSGs for this application

requires simultaneous achievement of low 1/f noise and successful addressing of issues involved in balance to horizontal fields, issues involved in additional noise sources such as vibration and temperature variations, and issues involved with simultaneously operating multiple TSGs.
To simultaneously address a number of concerns related to these issues we devised an attractive modification of the TSG. Many of the noise sources mentioned in the last section are less serious if the superconducting structures operate in zero field. Because a SQUID is not an absolute sensor of magnetic field, the feedback involved in the TSG keeps all of the SQUID magnetometers operating in the same field, but not necessarily in zero field. Be replacing the reference SQUID magnetometer with an absolute field magnetometer, such as a flux gate, we should be able to guarantee each of the magnetometers sits basically in zero average field. In principle as far as the noise goes, both SQUID magnetometers will see the same level from noise flux gate so this noise should cancel perfectly in the output. The degree to which this can be achieved needs to be determined experimentally and understood. This is the subject of a follow on contract.

5. Conclusions

Over the course of this contract, the potential brought on by the discovery of high Tc superconductivity for developing 77 K SQUIDs was largely demonstrated to be quite realizable. Challenges remaining are of the nature of developing a more mature industrial technology and of understanding and controlling certain critical properties for better performance, such as low frequency noise. The concept of the three SQUID gradiometer was demonstrated in high-Tc technology and should be an important part of the practical future of high-Tc naval gradiometers.

6. Contract Publications

1. J.Z. Sun, W.J. Gallagher, and R.H. Koch, "Magnetic Hysteresis in Thin Film dc SQUID Magnetometers," Applied Phys. Lett. **61**, 3190 (1992).

2. R. B. Laibowitz, R. P. Robertazzi, R. H. Koch, A. W. Kleinsasser, J. R. Kirtely, J. M. Viggiano, R. L Sandstrom, and W. J. Gallagher, "Tunnelling in all-high Tc edge junctions with deposited barriers", Phys. Rev. B, **46**, 14830 (1992).

3. J.Z. Sun, W.J. Gallagher, and R.H. Koch, "Non-linear hysteresis in Thin Film SQUID Magnetometers," IEEE Trans. Appl. Supercond. **3**, 2022 (1993).

4. R.P. Robertazzi, R.H. Koch, R.B. Laibowitz, and W.J. Gallagher, "Fabrication of YBCO/MgO/YBCO Micro-constriction Edge Junctions," IEEE Trans. Appl. Supercond. **3**, 2030 (1993).

5. R.P. Robertazzi and B..D. Oh, "Non-silver-paste method for making thermal contact to substrates for high-Tc film growth," IEEE Trans. on Superconductivity, **3**, 1094 (1993).

6. R.P. Robertazzi, A.W. Kleinsasser, R.B. Laibowitz, R.H. Koch, and K.G. Stawiaz, "Investigation of in-situ Ag/YBCO contacts for SNS devices," IEEE Trans. on Superconductivity, **3**, 1308 (1993).

7. R.H. Koch, J.R. Rozen, J.Z. Sun, and W.J. Gallagher, "Three SQUID Gradiometer," Applied Phys. Lett. **63**, 403 (1993).

8. J.Z. Sun, W.J. Gallagher, A.C. Callegari, V. Foglietti, and R.H. Koch, "Improved Process for high-Tc Step-Edge Junctions," Applied Phys. Lett. **63**, 1561 (1993).

9. J.Z. Sun, W.J. Gallagher, and R.H. Koch, "Improved Magnetic Hysteresis in Thin Film High-Tc SQUIDs," **Extended Abstracts of the 4th International Superconductive Electronics Conference**, (Boulder Colorado, August 11-14, 1993), pp. 152-153.

10. R.H. Koch, V. Foglietti, J.R. Rozen, J.Z. Sun, R.L. Sandstrom, and W.J. Gallagher "High-Tc Three-SQUID Gradiometer," **Extended Abstracts of the 4th International Superconductive Electronics Conference**, (Boulder Colorado, August 11-14, 1993), pp. 166-167.

11. G.M. Fisher, J. Mygind, N.F. Pedersen, and R.B. Laibowitz, "Linewidth Measurements in YBCO Junctions," **Extended Abstracts of the 4th International Superconductive Electronics Conference**, (Boulder Colorado, August 11-14, 1993), pp. 225-6.

12. W. Eidelloth, B. Oh, W.J. Gallagher, R.P. Robertazzi, and R.H. Koch, "$YBa_2Cu_3O_7$ Thin Film Gradiometers: Fabrication and Performance," Appl. Phys. Lett. **59**, 3473 (1991).

13. R.H. Koch, J.R. Rozen, J.Z. Sun, and W.J. Gallagher, "Three SQUID Gradiometer," Applied Phys. Lett. **63**, 403 (1993).

14. J.Z. Sun, W.J. Gallagher, A.C. Callegari, V. Foglietti, and R.H. Koch, "Improved Process for high-Tc Step-Edge Junctions," Applied Phys. Lett. **63**, 1561 (1993).

15. R.B. Laibowitz, J.Z. Sun, V. Foglietti, W.J. Gallagher, and R.H. Koch "High-Tc Multilevel Edge Junctions SQUIDs with $SrTiO_3$ Barriers Operating at 77 K," Applied Phys. Lett. **64**, 247 (1994).

16. R.H. Koch, V. Foglietti, J.R. Rozen, K.G. Stawiasz, M.B. Ketchen, J.Z. Sun, W.J. Gallagher, and D.K. Lathrop, "The effects of radio frequency radiation on the dc SQUID," Applied Phys. Lett. **65**, 100 (1994).

17. J.Z. Sun, W.J. Gallagher, and R.H. Koch, "Magnetic hysteresis in thin film SQUID magnetometers," Applied Phys. Lett. **61**, 3190 (1992).

18. J.Z. Sun, W.J. Gallagher, and R.H. Koch, "Initial-vortex-entry-related magnetic hysteresis in thin film SQUID magnetometers," Phys. Rev. B50, 13664, (1994).

19. V. Foglietti, R.H. Koch, J.Z. Sun, R.B. Laibowitz, and W.J. Gallagher, "Characterizing, Modelling, and Optimizing High-Tc SQUIDs," J. Appl. Phys. **77**, 378 (1995).

20. J.R. Kirtley, M.B. Ketchen, K.G. Stawiasz, J.Z. Sun, W.J. Gallagher, S.H. Blanton, and S.J. Wind, "High-resolution integrated scanning SQUID microscope", Appl. Phys. Lett. **66**, 1138 (1995).

21. J.Z. Sun, L.S. Yu-Jahnes, V. Foglietti, R.H. Koch, and W.J. Gallagher "Properties of YBaCuO Thin Film Single-Level dc SQUIDs Fabricated Using Step-Edge Junctions," IEEE Transactions on Superconductivity, **5**, 2107 (1995)

22. R.B. Laibowitz, J.Z. Sun, V. Foglietti, R.H. Koch, R.A. Altman and W.J. Gallagher "Properties of Multilevel Ramp Edge Junctions and SQUIDs with Laser-Ablated $SrTiO_3$ Barriers," IEEE Transactions on Superconductivity, **5**, 2620 (1995)

23. R.H. Koch, J. Z. Sun, V. Foglietti, and W. J. Gallagher, Flux Dam, A Method to Reduce Extra Low Frequency Noise When a Superconducting Magnetometer Is Exposed to a Magnetic Field, Appl. Phys. Lett., **67**, 709 (1995).

24. Bertha P. Chang, Neville Sonnenberg, and Michael J. Cima, Jonathan Z. Sun and Lock See Yu-Jahnes, Epitaxial Planarization of Patterned Yttria-Stabilized Zirconia Substrates for Multilayer Structures, Appl. Phys. Lett. **67**, 1148 (1995).

25. J.R. Kirtley, M.B. Ketchen, C.C. Tsuei, J.Z. Sun, W.J. Gallagher, Lock See Yu-Jahnes, A. Gupta, K.G. Stawiasz, and S.J. Wind, "Scanning SQUID microscopy", IBM Journal of Research and Development, 39, 655 (1995).

26. J. Z. Sun, W. J. Gallagher, and R. H. Koch, Direct-coupled high-Tc thin film SQUIDs using step-edge weak-link junctions, Applied Superconductivity, 13, 425 (1995).

27. Q.Y. Ma, S.H. Hong, J.R. Miller, J.Z. Sun, W.J. Gallagher, and R.B. Laibowitz, "High-Tc SQUICs Fabricated by Inhibition Ion Implantation," submitted to Superconducting Science and Technology, **9**, 92 (1996).

7. Patents

W. Eidelloth and W.J. Gallagher, “Selective Insulation Etching For Fabricating Superconductor Microcircuits”, United States Patent, 5,646,095 July 8, 1997.